Theory of spin precession monitored by laser pulse

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Abstract

We first predict the splitting of a spin degenerate impurity level when this impurity is irradiated by a circularly polarized laser beam tuned in the transparency region of a semiconductor. This splitting, which comes from different exchange processes between the impurity electron and the virtual pairs coupled to the pump beam, induces a spin precession around the laser beam axis, which lasts as long as the pump pulse. It can thus be used for ultrafast spin manipulation. This effect, which has similarities with the exciton optical Stark effect we studied long ago, is here derived using the concepts we developed very recently to treat many-body interactions between composite excitons and which make the physics of this type of effects quite transparent. They, in particular, allow to easily extend this work to other experimental situations in which a spin rotates under laser irradiation.

PACS.: 71.35.-y Excitons and related phenomena

Long ago, Danièle Hulin and her group [1] discovered that, when a semiconductor is irradiated by photons with energy too low to create electron-hole pairs, the exciton line blue-shifts. We have shown [2] that this shift, which disappears when the pump laser is turned off, comes from interactions between the real exciton created by the probe photon and the virtual excitons coupled to the pump beam [3].

In this communication, we predict an effect which has similarities with this exciton optical Stark shift: When an impurity is irradiated by a pump beam tuned in the transparency region of a semiconductor, its electronic levels shift: The electron bound to the donor interacts with the virtual electron-hole pairs coupled to the pump beam, either by Coulomb interaction, or by Pauli exclusion. If we choose the pump polarization in such a way that the exchange processes between the virtual pair and the up and down electrons of the impurity are different, this Pauli "interaction" splits the impurity level. As a result, the spin of the impurity electron precesses around the laser beam axis, as long as the pump is turned on. This effect can thus be used for ultrafast spin manipulation, a subject of great technological interest in the present days [4-10].

The impurity shift induced by a pump beam is derived following a procedure inspired from the one we used long ago to get the exciton optical Stark shift [3]. However, to enlighten the physics of this effect, we here calculate it using a "commutation technique" similar to the one we recently developed for excitons interacting with excitons [11] and which allows to identify the two basic ingredients of the electron-virtual pair interactions, namely a *direct* Coulomb scattering and a Pauli (or exchange) "scattering" — without any Coulomb contribution. The shift results from the interplay between the two, while the splitting only comes from different carrier exchanges.

To make the physics which controls the impurity level shift more transparent, we, in the first part, assume that the impurity electron and the electron of the virtual pairs have the same spin. The spin degrees of freedom and the laser polarization, of course crucial to get an impurity level splitting, will be introduced in the second part.

We end this communication by reconsidering other experimental conditions in which spins can rotate under laser pulse, namely free electron in a quantum well [8,9] and electrons trapped in quantum dots [5]. We explicitly show how our present theory can be easily extended to these cases.

Impurity level shift under laser irradiation

Let us consider a semiconductor having a ionized donor. Its Hamiltonian reads $H'_{sc} = H_{sc}+W_I$, where $H_{sc} = H_0+W_{sc}$ is the bare semiconductor Hamiltonian, with $H_0 = h_e+h_h$ and $W_{sc} = V_{ee}+V_{hh}+V_{eh}$, while W_I is the Coulomb interaction between the ionized donor and the carriers. This interaction, $\sum_n[-e^2/r_{e_n} + e^2/r_{h_n}]$, reads in second quantization,

$$W_I = -\sum_{\mathbf{k},\mathbf{q}} V_{\mathbf{q}}\, a^\dagger_{\mathbf{k}+\mathbf{q}}\, a_{\mathbf{k}} + \sum_{\mathbf{k},\mathbf{q}} V_{\mathbf{q}}\, b^\dagger_{\mathbf{k}+\mathbf{q}}\, b_{\mathbf{k}} \ . \tag{1}$$

$a^\dagger_{\mathbf{k}}$ and $b^\dagger_{\mathbf{k}}$ are the creation operators for free electrons and holes, *i. e.*, $(h_e - \epsilon^{(e)}_{\mathbf{k}})a^\dagger_{\mathbf{k}}|v\rangle = 0$, while $V_{\mathbf{q}} = 4\pi e^2/\mathcal{V}q^2$ or $2\pi e^2/\mathcal{S}q$ are the Coulomb matrix elements between free carriers, in 3D or 2D systems. In the presence of the ionized donor, the H'_{sc} one-electron eigenstates, $(H'_{sc} - \epsilon_\mu)|f_\mu\rangle = 0$, can be formally written as

$$|f_\mu\rangle = a^\dagger_\mu|v\rangle = \sum_{\mathbf{k}} \langle\mathbf{k}|f_\mu\rangle a^\dagger_{\mathbf{k}}|v\rangle \ . \tag{2}$$

If we now irradiate this system with pump photons $(\omega_0, \mathbf{Q}_0)$, the coupled matter-photon Hamiltonian reads $\mathcal{H} = H'_{sc} + H_{ph} + \mathcal{U}$, where $H_{ph} = \omega_0 c^\dagger_0 c_0$ is the bare photon Hamiltonian and $\mathcal{U} = (U^\dagger_0 c_0 + h.c.)$ the semiconductor-photon coupling. $U^\dagger_0$, which creates one electron-hole pair with momentum $\mathbf{Q}_0$, can be written as $U^\dagger_0 = A^* \sum_{\mathbf{p}} B^\dagger_{\mathbf{p},\mathbf{Q}_0}$ where $B^\dagger_{\mathbf{p},\mathbf{Q}} = a^\dagger_{\mathbf{p}+\alpha_e\mathbf{Q}}\, b^\dagger_{-\mathbf{p}+\alpha_h\mathbf{Q}}$, with $\alpha_e = 1 - \alpha_h = m_e/(m_e + m_h)$, is the creation operator for one free electron-hole pair [12] with center of mass momentum $\mathbf{Q}$ and relative motion momentum $\mathbf{p}$. It is such that $(H_0 - E_g - E_{\mathbf{p},\mathbf{Q}})B^\dagger_{\mathbf{p},\mathbf{Q}}|v\rangle = 0$, where E_g is the band gap, while $E_{\mathbf{p},\mathbf{Q}} = \hbar^2\mathbf{p}^2/2m + \hbar^2\mathbf{Q}^2/2M$, with $m^{-1} = m_e^{-1} + m_h^{-1}$ and $M = m_e + m_h$, is the $(\mathbf{p},\mathbf{Q})$ pair energy.

As for the exciton optical Stark effect [3], the impurity level shift results from the difference between the impurity level change and the vacuum level change induced by the pump beam. For $\mathcal{U} = 0$, the eigenstate with a ionized impurity and N_0 photons is $|v\rangle \otimes |N_0\rangle$, its energy being $\mathcal{E}_0 = N_0\omega_0$. At lowest order in $\mathcal{U}$, this energy becomes

$$\mathcal{E}'_0 \simeq N_0\omega_0 + N_0\langle v|U_0 \frac{1}{\omega_0 - H'_{sc}} U^\dagger_0|v\rangle \ . \tag{3}$$

In a similar way, for $\mathcal{U} = 0$, the eigenstates with one electron and N_0 photons are $a^\dagger_\mu|v\rangle \otimes |N_0\rangle$, their energy being $\mathcal{E}_\mu = \epsilon_\mu + N_0\omega_0$, while at lowest order in $\mathcal{U}$, they read

$$\mathcal{E}'_\mu \simeq \epsilon_\mu + N_0\omega_0 + N_0\langle v|a_\mu\, U_0 \frac{1}{\omega_0 + \epsilon_\mu - H'_{sc}} U^\dagger_0\, a^\dagger_\mu|v\rangle \ , \tag{4}$$

The impurity level shift induced by the pump beam, $[\mathcal{E}'_\mu-\mathcal{E}'_0]-[\mathcal{E}_\mu-\mathcal{E}_0]$, is thus

$$\Delta_\mu = N_0\langle v|U_0\left(a_\mu\,\frac{1}{\omega_0+\epsilon_\mu-H'_{sc}}\,a_\mu^\dagger\;-\;\frac{1}{\omega_0-H'_{sc}}\right)U_0^\dagger|v\rangle\ . \tag{5}$$

This quantity, linear in the pump intensity, is formally similar to our expression of the exciton optical Stark shift, with $a_\mu^\dagger$ and ϵ_μ just replacing the probe exciton creation operator $B_t^\dagger$ and energy E_t.

In order to calculate Δ_μ, we introduce the Coulomb *creation* potential $V_\mu^\dagger$, which, in this problem, is defined as $[H'_{sc},a_\mu^\dagger]=\epsilon_\mu a_\mu^\dagger+V_\mu^\dagger$. It precisely reads

$$V_\mu^\dagger=\sum_{\mathbf{k}}\langle\mathbf{k}|f_\mu\rangle\sum_{\mathbf{q}}V_{\mathbf{q}}\,a_{\mathbf{k}+\mathbf{q}}^\dagger\sum_{\mathbf{k}'}\left(a_{\mathbf{k}'-\mathbf{q}}^\dagger\,a_{\mathbf{k}'}-b_{\mathbf{k}'-\mathbf{q}}^\dagger\,b_{\mathbf{k}'}\right)\ . \tag{6}$$

From the formal definition of $V_\mu^\dagger$, it is easy to check that

$$\frac{1}{x-H'_{sc}}\,a_\mu^\dagger=a_\mu^\dagger\,\frac{1}{x-H'_{sc}-\epsilon_\mu}+\frac{1}{x-H'_{sc}}\,V_\mu^\dagger\,\frac{1}{x-H'_{sc}-\epsilon_\mu}\ , \tag{7}$$

which is valid for any scalar x. This allows to split Δ_μ as

$$\Delta_\mu=N_0\left[\frac{1}{2}(\alpha_\mu+\alpha_\mu^*)+\frac{1}{2}(\beta_\mu+\beta_\mu^*)+\gamma_\mu\right]\ . \tag{8}$$

Note that we have done a similar splitting in the case of the exciton optical Stark shift [3].

As explicitly shown below, α_μ, given by

$$\alpha_\mu=\langle v|U_0\,a_\mu^\dagger a_\mu\,|\psi_0\rangle\ , \tag{9}$$

in which we have set $|\psi_0\rangle=(H'_{sc}-\omega_0)^{-1}U_0^\dagger|v\rangle$, comes from "Pauli interaction" between the impurity electron and the virtual pair. On the opposite, β_μ and γ_μ, given by

$$\begin{aligned}\beta_\mu&=\langle\psi_0|a_\mu\,V_\mu^\dagger|\psi_0\rangle\\ \gamma_\mu&=\langle\psi_0|V_\mu\,(\omega_0+\epsilon_\mu-H'_{sc})^{-1}\,V_\mu^\dagger|\psi_0\rangle\ ,\end{aligned} \tag{10}$$

contain one or two $V_\mu^\dagger$ operators, so that they come from Coulomb interaction between the impurity electron and the virtual pair.

In the following, it will be convenient to develop $|\psi_0\rangle$ on free pair states, according to

$$|\psi_0\rangle=A^*\sum_{\mathbf{p},\mathbf{Q}}G(\mathbf{p},\mathbf{Q})\,B_{\mathbf{p},\mathbf{Q}}^\dagger|v\rangle\ , \tag{11}$$

$$G(\mathbf{p},\mathbf{Q}) = \sum_{\mathbf{p}'} \langle v| B_{\mathbf{p},\mathbf{Q}} \, \frac{1}{H'_{sc} - \omega_0} \, B^\dagger_{\mathbf{p}',\mathbf{Q}_0} |v\rangle \ . \tag{12}$$

"Commutation technique" for a free pair interacting with an impurity electron

α_μ and β_μ are easy to write in terms of the two "scatterings" controling the physics of this problem, namely Λ and Ξ^{dir}, which appear in a "commutation technique" inspired from the one we recently developed to treat many-body effects between composite excitons [11]. From

$$\left[a_{\mu'} a^\dagger_\mu \ , B^\dagger_{\mathbf{p},\mathbf{Q}} \right] = - \sum_{\mathbf{p}',\mathbf{Q}'} \Lambda_{\mu'\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}} \, B^\dagger_{\mathbf{p}',\mathbf{Q}'} \ , \tag{13}$$

one of these two "scatterings", which is dimensionless, is found to be

$$\begin{aligned} \Lambda_{\mu'\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}} &= \langle f_{\mu'}|\mathbf{p} + \alpha_e\mathbf{Q}\rangle \, \langle \mathbf{p}' + \alpha_e\mathbf{Q}'|f_\mu\rangle \, \delta_{-\mathbf{p}'+\alpha_h\mathbf{Q}',-\mathbf{p}+\alpha_h\mathbf{Q}} \\ &\equiv \int d\mathbf{r}_e \, d\mathbf{r}_{e'} \, d\mathbf{r}_h \, \langle f_{\mu'}|\mathbf{r}_e\rangle \, \langle \mathbf{p}',\mathbf{Q}'|\mathbf{r}_{e'},\mathbf{r}_h\rangle \, \langle \mathbf{r}_e,\mathbf{r}_h|\mathbf{p},\mathbf{Q}\rangle \, \langle \mathbf{r}_{e'}|f_\mu\rangle \ . \end{aligned} \tag{14}$$

It corresponds to a bare electron exchange between the impurity level μ and the pair $(\mathbf{p},\mathbf{Q})$, which transforms them into an "out" impurity level μ' and an "out" pair $(\mathbf{p}',\mathbf{Q}')$. Note that this Λ scattering is Coulomb free. From eq. (13), we can show that

$$\langle v|B_{\mathbf{p}',\mathbf{Q}'} \, a_{\mu'} \, a^\dagger_\mu \, B^\dagger_{\mathbf{p},\mathbf{Q}}|v\rangle = \delta_{\mu',\mu} \, \delta_{\mathbf{p}',\mathbf{p}} \, \delta_{\mathbf{Q}',\mathbf{Q}} - \Lambda_{\mu'\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}} \ . \tag{15}$$

The second scattering, defined through

$$\left[V^\dagger_\mu, B^\dagger_{\mathbf{p},\mathbf{Q}} \right] = \sum_{\mu',\mathbf{p}',\mathbf{Q}'} \Xi^{\mathrm{dir}}_{\mu'\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}} \, a^\dagger_{\mu'} \, B^\dagger_{\mathbf{p}',\mathbf{Q}'} \ , \tag{16}$$

is found to be

$$\begin{aligned} \Xi^{\mathrm{dir}}_{\mu'\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}} &= V_{\mathbf{Q}-\mathbf{Q}'} \sum_{\mathbf{k},\mathbf{k}'} \langle f_{\mu'}|\mathbf{k}'\rangle \, \langle \mathbf{k}|f_\mu\rangle \, \delta_{\mathbf{k}'+\mathbf{Q}',\mathbf{k}+\mathbf{Q}} \left[\delta_{-\mathbf{p}'+\alpha_h\mathbf{Q}',-\mathbf{p}+\alpha_h\mathbf{Q}} - \delta_{\mathbf{p}'+\alpha_e\mathbf{Q}',\mathbf{p}+\alpha_e\mathbf{Q}} \right] \\ &\equiv \int d\mathbf{r}_e \, d\mathbf{r}_{e'} \, d\mathbf{r}_h \, \langle f_{\mu'}|\mathbf{r}_{e'}\rangle \, \langle \mathbf{p}',\mathbf{Q}'|\mathbf{r}_e,\mathbf{r}_h\rangle [v_{e'e} - v_{e'h}] \, \langle \mathbf{r}_e,\mathbf{r}_h|\mathbf{p},\mathbf{Q}\rangle \, \langle \mathbf{r}_{e'}|f_\mu\rangle \ , \end{aligned} \tag{17}$$

where $v_{ij} = e^2/|\mathbf{r}_i - \mathbf{r}_j|$. It corresponds to *direct* Coulomb interactions between the impurity electron and the pair, *without* any carrier exchange.

Calculation of the impurity level shift

(i) *Pure Pauli term*

Equations (9,11,15) allow to write α_μ as

$$\alpha_\mu = |A|^2 \sum_{\mathbf{p}',\mathbf{p},\mathbf{Q}} \Lambda_{\mu\mathbf{p}'\mathbf{Q}_0;\mu\mathbf{p}\mathbf{Q}} \, G(\mathbf{p},\mathbf{Q}) \ . \tag{18}$$

Equation (18) makes clear that this part of the shift is linked to "Pauli interaction", *i. e.*, exchange between the impurity electron and the virtual pairs. By noting that, at large detuning $\Omega = E_g - \omega_0$, $G(\mathbf{p},\mathbf{Q})$ tends to $\delta_{\mathbf{Q},\mathbf{Q}_0}/\Omega$, we can extract this limit from α_μ to write it as $\alpha_\mu = |A|^2(1+\eta)/\Omega$, where, due to eqs. (12,14), η is precisely given by

$$\eta = \sum_{\mathbf{p}',\mathbf{p},\mathbf{Q}} \langle \mathbf{p} - \alpha_h \mathbf{Q} + \mathbf{Q}_0 | f_\mu \rangle \langle f_\mu | \mathbf{p} + \alpha_e \mathbf{Q} \rangle \langle v | B_{\mathbf{p},\mathbf{Q}} \left[\frac{E_g - \omega_0}{H'_{sc} - \omega_0} - 1 \right] B^\dagger_{\mathbf{p}',\mathbf{Q}_0} | v \rangle \ . \qquad (19)$$

For large Ω, the bracket of eq. (19) tends to zero, so that α_μ does reduce to $|A|^2/\Omega$. This limit has to be compared to the one of the similar Pauli term α in the optical Stark shift, namely $2|A|^2/\Omega$. The link between these two limits can be physically understood by noting that, in the case of the exciton shift, a virtual pump pair can exchange both, its electron and its hole, with the probe exciton, while here, it can only exchange its electron with the impurity level: The numerical prefactor of the large detuning leading term just results from one carrier exchange instead of two.

In order to calculate the next order term η, we use

$$\frac{1}{\omega_0 - H'_{sc}} = \frac{1}{\omega_0 - H_0} + \frac{1}{\omega_0 - H_0}(W_I + W_{sc})\frac{1}{\omega_0 - H_0} + \cdots \ , \qquad (20)$$

which follows from $H'_{sc} = H_0 + W_I + W_{sc}$. The first term of eq. (20) leads to replace H'_{sc} by H_0 in eq. (19). As $Q_0 \ll 1/a_X$, while for bound states $|\langle \mathbf{k} | f_\mu \rangle|^2 \simeq 0$ for $k \gg 1/a_X$, we find that the contribution of this term to η is of the order of R_X/Ω, where $R_X = \hbar^2/2ma_X^2$. A similar R_X/Ω behavior is found for each of the two terms of W_I. On the opposite, the W_{sc} term of eq. (20), which corresponds to Coulomb interaction *inside* the virtual pair (see fig. 1b), becomes singular for large momentum transfers. It leads to

$$\eta \simeq \Omega \sum_{\mathbf{p},\mathbf{p}'} \frac{|\langle \mathbf{p} + \alpha_e \mathbf{Q}_0 | f_\mu \rangle|^2 \, V_{\mathbf{p}'-\mathbf{p}}}{(\Omega + E_{\mathbf{p},\mathbf{Q}_0})(\Omega + E_{\mathbf{p}',\mathbf{Q}_0})} \simeq \sum_{\mathbf{p}'} \frac{V_{\mathbf{p}'}}{\Omega + \hbar^2 \mathbf{p}'^2/2m} = \tilde{\alpha} \sqrt{\frac{R_X}{\Omega}} \ , \qquad (21)$$

with $\tilde{\alpha} = 2$ for 3D and $\tilde{\alpha} = \pi$ for 2D; so that we end with

$$\alpha_\mu = \frac{|A|^2}{\Omega} \left[1 + \tilde{\alpha} \sqrt{\frac{R_X}{\Omega}} + O\left(\frac{R_X}{\Omega}\right) \right] \ . \qquad (22)$$

Note that, as $R_X \propto e^4$, $\sqrt{R_X/\Omega}$ is in fact the dimensionless parameter associated to a Coulomb expansion.

(ii) *First order Coulomb term between the impurity electron and the virtual pairs*

We now turn to β_μ. Using eqs. (10,11,15,16), it reads

$$\beta_\mu = |A|^2 \sum_{\mathbf{p}',\mathbf{Q}',\mathbf{p},\mathbf{Q}} G^*(\mathbf{p}',\mathbf{Q}') \left[\Xi^{\mathrm{dir}}_{\mu\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}} - \Xi^{\mathrm{in}}_{\mu\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}} \right] G(\mathbf{p},\mathbf{Q}) \ , \tag{23}$$

where $\Xi^{\mathrm{in}}_{\mu'\mathbf{p}'\mathbf{Q}';\mu\mathbf{p}\mathbf{Q}}$ is the sum over $(\mu'',\mathbf{p}'',\mathbf{Q}'')$ of $\Lambda_{\mu'\mathbf{p}'\mathbf{Q}';\mu''\mathbf{p}''\mathbf{Q}''}\, \Xi^{\mathrm{dir}}_{\mu''\mathbf{p}''\mathbf{Q}'';\mu\mathbf{p}\mathbf{Q}}$. Being made of a direct Coulomb process *between* the impurity electron and the pair, followed by an electron exchange (see fig. 1c), Ξ^{in} is actually an exchange Coulomb scattering.

To get the β_μ lowest order term in $\sqrt{R_X/\Omega}$, *i. e.*, in Coulomb interaction, we can replace H'_{sc} by its free carrier expression H_0, *i. e.*, $G(\mathbf{p},\mathbf{Q})$ by $\delta_{\mathbf{Q},\mathbf{Q}_0}/(\Omega+E_{\mathbf{p},\mathbf{Q}})$. The two terms of Ξ^{dir} being then equal, we are left with the exchange term, which gives

$$\begin{aligned} \beta_\mu &\simeq |A|^2 \sum_{\mathbf{k},\mathbf{p}} |\langle \mathbf{k}|f_\mu\rangle|^2 \frac{V_{\mathbf{p}+\alpha_e\mathbf{Q}_0-\mathbf{k}}}{\Omega+E_{\mathbf{p},\mathbf{Q}_0}} \left[-\frac{1}{\Omega+E_{\mathbf{p},\mathbf{Q}_0}} + \frac{1}{\Omega+E_{\mathbf{k}-\alpha_e\mathbf{Q}_0,\mathbf{Q}_0}} \right] \\ &\simeq \frac{|A|^2}{\Omega} \left[\frac{\tilde{\alpha}}{2} \sqrt{\frac{R_X}{\Omega}} + O\left(\frac{R_X}{\Omega}\right) \right] \ . \end{aligned} \tag{24}$$

(iii) *Correlation term*

The last contribution γ_μ contains two Coulomb interactions between the impurity electron and the free pair, *i. e.*, two ($e^2 \propto \sqrt{R_X}$) factors. In the large detuning limit, it thus behaves as R_X/Ω at least (other e^2 factors possibly appearing if we expand $(\omega_0+\epsilon_\mu - H'_{sc})^{-1}$ according to eq. (7)). Consequently, in this large detuning limit, γ_μ is negligible in front of α_μ and β_μ. On the opposite, the γ_μ contribution is the one possibly leading to resonances in the impurity level shift. Indeed, if we look at eq. (10), we see that γ_μ contains $(\omega_0+\epsilon_\mu - H'_{sc})^{-1}$ acting on two electrons plus one hole. The corresponding H'_{sc} eigenstates being the excitons bound to an impurity, we can inject the closure relation for these states in front of this H'_{sc} dependent operator. γ_μ then shows poles at $\omega_0 = E_g + \hat{\epsilon}_\mu$, where the $\hat{\epsilon}_\mu$'s are the energies of these excitons bound on impurity.

This leads us to conclude that, at large detuning, the impurity level shift $|A|^2 N_0/\Omega$ is entirely controlled by electron exchange between the impurity and the virtual pairs coupled to the pump beam, without any Coulomb contribution (see fig. 1a). The next order term, which is $\sqrt{R_X}/\Omega$ smaller, is also due to an electron exchange but contains, in addition, one Coulomb interaction, either inside the virtual pairs as in α_μ (see fig. 1b), or between these virtual pairs and the impurity electron as in β_μ (see fig. 1c). On the opposite, possible resonances at the bound exciton energies can be found in the correlation term γ_μ, which, at large detuning, gives a negligible contribution.

Impurity level splitting

Let us now see how the pump polarization and the spin degrees of freedom affect these results.

The semiconductor-photon coupling now reads

$$U_0^\dagger = \sum_{\mathbf{p},s,m} A_{s,m}^* \, B_{\mathbf{p},\mathbf{Q}_0;s,m}^\dagger \ , \tag{25}$$

where $B_{\mathbf{p},\mathbf{Q};s,m}^\dagger = a_{\mathbf{p}+\alpha_e\mathbf{Q},s}^\dagger b_{-\mathbf{p}+\alpha_h\mathbf{Q},m}^\dagger$ creates a pair with an electron spin $s = \pm 1/2$ and a hole momentum $m = (\pm 3/2, \pm 1/2)$ for bulk materials while $m = (\pm 3/2)$ only for quantum wells. The $A_{s,m}$'s depend on photon polarization. For bulk materials, their non-zero values are $A_{\mp 1/2,\pm 3/2} = A_\pm$ and $A_{\pm 1/2,\pm 1/2} = -A_\pm/\sqrt{3}$, while, for quantum wells, these $A_{\pm 1/2,\pm 1/2}$'s are zero . In the case of a circularly polarized beam $\sigma_\pm$, the $A_\pm$'s are such that $A_\pm = A$ and $A_\mp = 0$, while for a linear beam along x (resp. y), they are $A_+ = A_- = A/\sqrt{2}$ (resp. $A_+ = -A_- = A/\sqrt{2}$).

In addition to these complexities in the semiconductor-photon interaction, we have also to take into account the fact that the impurity levels are now degenerate, the up and down spins having the same energy — in the absence of pump beam. Consequently, it is now necessary to use degenerate perturbation theory to get the impurity level change induced by the laser beam. It is possible to show that this change is obtained from the diagonalization of a 2×2 matrix, its eigenvalues being

$$\mathcal{E}'_\mu = \epsilon_\mu + N_0\omega_0 + \frac{N_0}{2}\left[d_{++} + d_{--} \pm \sqrt{(d_{++} - d_{--})^2 + 4|d_{+-}|^2}\right] \ , \tag{26}$$

$$d_{\sigma'\sigma} = \langle v|U_0 \, a_{\mu,\sigma'} \, \frac{1}{\omega_0 + \epsilon_\mu - H'_{sc}} \, a_{\mu,\sigma}^\dagger \, U_0^\dagger|v\rangle \ . \tag{27}$$

By taking into account the vacuum level change induced by the pump beam, still given by eq. (3), we end with an impurity level having an average shift equal to $\hat{\Delta} = N_0(\hat{d}_{++} + \hat{d}_{--})/2$, and a splitting given by $\hat{\delta} = N_0(\sqrt{(\hat{d}_{++} - \hat{d}_{--})^2 + 4|\hat{d}_{+-}|^2}$, where

$$\hat{d}_{\sigma'\sigma} = d_{\sigma'\sigma} - \delta_{\sigma',\sigma}\langle v|U_0(\omega_0 - H'_{sc})^{-1}U_0^\dagger|v\rangle \ . \tag{28}$$

Note that eq. (28) is a generalization of eq. (5), in the presence of spin degrees of freedom.

To get these $\hat{d}_{\sigma'\sigma}$, we use a commutation technique similar to the one without spin. In the presence of spins, the three scatterings Ξ^{dir}, Ξ^{in} and Λ are now the product of an orbital part, which is the one without spin, and a spin part. Due to spin conservation

in Coulomb and exchange processes, this spin part is just $\delta_{\sigma',\sigma}\delta_{s',s}\delta_{m',m}$ for the direct scattering $\Xi^{\rm dir}$ (see fig. (1d), and $\delta_{\sigma',s}\delta_{s',\sigma}\delta_{m',m}$ for the exchange scatterings $\Xi^{\rm in}$ and Λ (see fig. 1e).

It is then easy to show that, again, the large detuning leading term of $\hat{d}_{\sigma'\sigma}$ is entirely controlled by electron exchange between the impurity and the virtual pairs coupled to the pump, the next order term having just one additional Coulomb interaction either inside the pair or between the pair and the impurity electron. The two first terms of $N_0\hat{d}_{\sigma'\sigma}$ correspond to the two first terms of Δ_μ as obtained previously in eqs. (18) and (23), with $|A|^2$ just replaced by

$$\pi_{\sigma'\sigma} = \sum_m A^*_{\sigma',m}\, A_{\sigma,m} \ . \tag{29}$$

We can then note that $\pi_{+-} = 0$, since for a given m, there is only one σ which makes $A_{\sigma,m} \neq 0$, while $\pi_{\pm\pm}$ is equal to $|A_\mp|^2 + |A_\pm|^2/3$ for bulk samples, and $|A_\mp|^2$ for quantum wells. This shows that, when the pump beam is linear, $|A_+| = |A_-|$ so that $\pi_{++} = \pi_{--}$: The impurity level has a blue shift equal to $\Delta_\mu/2$ for quantum wells, and $2\Delta_\mu/3$ for bulk materials, but no splitting. On the opposite, for circular beams, $A_+A_- = 0$, so that the impurity level splits: One impurity level blue shifts of an amount Δ_μ, while the other is unchanged for quantum wells, or shifted by $\Delta_\mu/3$ for bulk materials. The splitting $\hat{\delta}$ is then either Δ_μ or $2\Delta_\mu/3$.

Spin precession of an impurity electron induced by a pump beam

Let us take $|\phi_0\rangle = (\cos\theta\, a^\dagger_{\mu+} + \sin\theta\, a^\dagger_{\mu-})|v\rangle$ as initial impurity state. If we turn on a circularly polarized pump beam which propagates along z, the up and down spins are shifted differently, due to their different electron exchanges with the virtual pairs, so that $|\phi_0\rangle$ becomes

$$|\phi_t\rangle = (\cos\theta\, a^\dagger_{\mu+} + e^{i\hat{\delta}t/\hbar}\, \sin\theta\, a^\dagger_{\mu-})|v\rangle \ , \tag{30}$$

within a phase factor, $\hat{\delta}$ being the shift between the $(\pm 1/2)$ impurity electrons calculated previously. The projections of $|\phi_t\rangle$ over $(+1/2)$ and $(-1/2)$ staying unchanged, the spin of the impurity electron thus precesses around the z axis with a period $T = 2\pi\hbar/\hat{\delta}$. Since $\hat{\delta}$ is of the order of Δ_μ — which is just the exciton optical Stark shift, within a factor 1/2, in the large detuning limit — , we thus expect a precession period of the order of 1psec within the experimental conditions giving an exciton optical Stark shift of the order of

1mev. We can note that this period is far shorter than the spin relaxation time, which is of the order of 1nsec.

Extension of the theory to other spin precessions

Let us end this communication by considering two cases in which spin precession induced by laser beams has been described.

(i) *Free electron in a quantum well* [8,9]

This case can be readily deduced from the above results by setting the Coulomb potential between the carriers and the ionized impurity W_I equal to zero. This leads to replace $a^\dagger_\mu$ by $a^\dagger_{\mathbf{k}_0}$, ϵ_μ by $\epsilon^{(e)}_{\mathbf{k}_0}$ and $|f_\mu\rangle$ by $|\mathbf{k}_0\rangle$, with $\langle\mathbf{k}|\mathbf{k}_0\rangle = \delta_{\mathbf{k},\mathbf{k}_0}$, in the formal expression of the shift Δ_μ as well as in its α_μ, β_μ and γ_μ contributions. We have shown that, in the large detuning limit, the two first terms of the shift are controlled by an electron exchange, with possibly one Coulomb interaction inside the virtual pairs or between these pairs and the impurity electron, the Coulomb interaction with the ionized donor entering at the next order, R_X/Ω, only. This shows that the shift and splitting of the impurity electron and the ones of a free electron are thus just the same for these two large detuning terms, provided that $\epsilon^{(e)}_{\mathbf{k}_0} \ll \Omega$, for eq. (21) to be valid. On the opposite, the possible resonances coming from the γ_μ contribution differ. They are now controlled by the two electron-one hole eigenstates, *i. e.*, the trions, while, in the presence of impurity, they are controlled by excitons bound to the impurity. As the coupling between photon and trion is in fact extremely weak in the large sample limit [13], the weights of these resonances are expected to be rather small.

(ii) *Electron in a quantum dot* [5]

The spin precession of an electron trapped in a quantum dot can also be deduced from the above theory. The one-body electron Hamiltonian h_e has just to now include the dot confinement. Instead of $a^\dagger_{\mathbf{k}}$, the creation operator for a Coulomb free electron reads $a^\dagger_n$, with $(h_e - \epsilon^{(e)}_n)\, a^\dagger_n|v\rangle = 0$. These eigenstates *a priori* include bound states as well as extended states, if the barrier height is finite.

If we now consider one electron trapped in the dot ground state $a^\dagger_{n_0}|v\rangle$, its shift Δ_{n_0} is given by eq. (5), with $a^\dagger_\mu$ and ϵ_μ replaced by $a^\dagger_{n_0}$ and $\epsilon^{(e)}_{n_0}$, while the dot-photon coupling has now to be written as $U^\dagger_0 = \sum_{n,m} A^*_{nm} a^\dagger_n b^\dagger_m$. This shift can be calculated using a

“commutation technique” formally similar, the Coulomb *creation* potential now reading

$$V_{n_0}^{\dagger}=\sum_{n} a_n^{\dagger} \sum_{m',m} \left[V_{ee}\left(\begin{smallmatrix} n & n_0 \\ m' & m \end{smallmatrix}\right) a_{m'}^{\dagger} a_m + V_{eh}\left(\begin{smallmatrix} n & n_0 \\ m' & m \end{smallmatrix}\right) b_{m'}^{\dagger} b_m\right] \ , \tag{31}$$

where $V_{ee}\begin{pmatrix} n' & n \\ m' & m \end{pmatrix}$ and $V_{eh}\begin{pmatrix} n' & n \\ m' & m \end{pmatrix}$ are the Coulomb matrix elements between dot states (n,m) and (n',m').

The calculation of Δ_{n0} which is performed in a quite similar way, shows that, at large detuning, the shift is again controlled by electron exchange, its leading term now reading $N_0\Omega^{-1}\sum_m |A_{n_0m}|^2$, while resonant contributions in the correlation term γ_μ must appear at the eigenenergies of a “trion” in the dot.

Conclusion

We have shown that, due to carrier exchanges between the impurity and the virtual pairs coupled to a pump beam tuned in the transparency region of a semiconductor, the up and down electronic levels of an impurity blue shift. The degenerate levels of this impurity can also split if the pump beam is circularly polarized, due to differences in these carrier exchanges. This splitting induces a spin precession around the laser beam axis, which lasts as long as the pulse. It can thus be used to manipulate spins. We have also shown how the present theory can be extended to other spin precessions induced by laser beam, such as the one of free electrons in a quantum well or the one of electrons trapped in a quantum dot.

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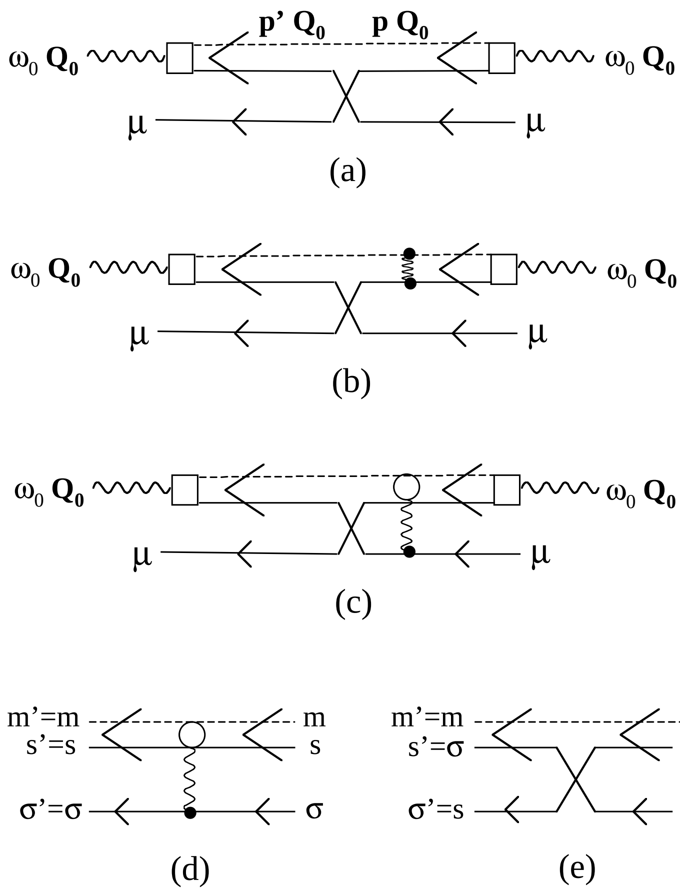

Figure 1: (a): A photon ($\omega_0\mathbf{Q}_0$) creates a virtual electron-hole pair ($\mathbf{p},\mathbf{Q}_0$). This pair exchanges its electron with the electron of an impurity (in a μ state) and finally recombines to give back the ($\omega_0\mathbf{Q}_0$) photon. This process is the dominant one in the impurity level shift at large detuning. (b,c): The large detuning next order term contains one Coulomb interaction either inside the virtual pair (b) or between this pair and the impurity electron (c). (d): The direct Coulomb scattering Ξ^{dir} of the "commutation technique" for a free pair interacting with an impurity electron: The "in" and "out" pairs are made with the same electron. (e): Exchange or Pauli "scattering" Λ of this commutation technique. Note that this scattering exists in the absence of any Coulomb process.